

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35.U.S.C. 371

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EK 839 854 188 US

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July 30, 2001

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A34514-PCT-USA - 072035.0128

U S APPLICATION NO
09/890431

INTERNATIONAL APPLICATION NO
PCT/GB00/00268

INTERNATIONAL FILING DATE
31 January 2000

PRIORITY DATE CLAIMED
1 February 1999

TITLE OF INVENTION
ELECTROLUMINESCENT MATERIALS

APPLICANT(S) FOR DO/EO/US
Poopathy Kathirgamanathan

Applicant herewith submits to the United States Designated /Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(I).
4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☒ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ A copy of the International Search Report (PCT/ISA/210)
 - a. ☒ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☐ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern other document(s) or information included:

11. ☒ A copy of the International Preliminary Examination Report (PCT/IPEA/409)
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A FIRST preliminary amendment.
14. ☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information:
 - a. ☒ a copy of the International Search Report (PCT/ISA/210)
 - b. ☒ a copy of the International Preliminary Examination Report (PCT/IPEA/409)
 - c. ☒ PCT application No. PCT/GB00/00268 was published in English under publication number WO 00/44851 on August 3, 2000.

INTERNATIONAL APPLICATION NO.
PCT/GB00/0268 09/890431INTERNATIONAL FILING DATE
31 January 2000PRIORITY DATE CLAIMED
1 February 1999

17. [X] The following fees are submitted:

Basic National Fee (37 CFR 1.492(a)(1)-(5):

Neither international preliminary examination fee (37 CFR 1.482)

Nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO (1.492(a)(3)) \$1,000.00

International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO (1.492(a)(5)) \$860.00

International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO (1.492(a)(2)) \$710.00

International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) (1.492(a)(1)) \$690.00

International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00

ENTER APPROPRIATE BASIC FEE AMOUNT = \$860.00

Surcharge of \$130.00 for furnishing the oath or declaration later than [] 20 [] 30 months from the earliest claimed priority date (37 C.F.R. 1.492(e)).

\$0.00

Claims	Number Filed	Number Extra	Rate	\$	
Total Claims	28 -20=	8	X \$ 18.00	\$144.00	
Independent Claims	2 -3=	0	X \$ 80.00	\$	
Multiple dependent claim(s) (if applicable)			+ \$270.00	\$	
TOTAL OF ABOVE CALCULATIONS =				\$1004.00	
Reduction by 1/2 for filing by small entity, if applicable				\$	
SUBTOTAL =				\$	
Processing fee of \$130.00 for furnishing the English translation later than [] 20 [] 30 months from the earliest claimed priority date (37 CFR 1.492(f)). +				\$	
TOTAL NATIONAL FEE =				\$1004	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +				\$ 40.00	
TOTAL FEES ENCLOSED =				\$1044.00	
				Amt. refunded	\$
				charged	\$1044

a. [X] A check in the amount of \$1044.00 to cover the above fees is enclosed.

b. [] Please charge our Deposit Account No. 02-4377 in amount of \$ ___ to cover the above fees. A copy of this sheet is enclosed.

c. [X] The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 02-4377. A copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

BAKER BOTTS L.L.P.
30 Rockefeller Plaza
New York, New York 10112-4498

Signature

July 30, 2001

Date

32,689

Registration No.

A34514-PCT-USA - 072035.0128

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : Poopathy Kathirgamanathan
Serial No. : Not Yet Known
Filed : Not Yet Known
For : ELECTROLUMINESCENT MATERIALS

PRELIMINARY AMENDMENT

I hereby certify that this paper is being deposited with the United States Postal Service as Express Mail in an envelope addressed to: Assistant Commissioner for Patents, Washington, D.C. 20231

July 30, 2001
Date of Deposit

Marta E. Delsignore
Attorney Name

Marta E. Delsignore
Signature

32,689
PTO Registration No.

July 30, 2001
Date of Signature

EXPRESS MAIL NO.: EK 839 854 188 US

Assistant Commissioner for Patents
Washington, D.C. 20231

Sir:

Preliminary to Examination, please amend the above-identified patent application as follows.

IN THE SPECIFICATION:

Page 1, before the first full paragraph please insert:

--This application is a national stage application of PCT/GB00/00268 which was published in English under publication number WO 00/44851 on August 3, 2001--

09/890431

A34514-PCT-USA - 072035.0128

PATENT

Page 1, after line 3, please insert:

--The present invention relates to electroluminescent materials which can emit light in the ultra-violet region of the spectrum and devices made using such materials.

BACKGROUND OF THE INVENTION--

Page 2, after line 13, please insert:

--For example, there are devices and displays etc. where ultra-violet light is used to excite other materials to cause these other materials to fluoresce in the visible spectrum.

SUMMARY OF THE INVENTION--

Page 4, after line 10, please insert:

--The materials of the present invention can be incorporated into electroluminescent devices which emit ultra-violet light and the invention include such electroluminescent devices.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 is a spectrum of gadolinium ethylenediaminetetracetate sodium salt; and

Figure 2 is a spectrum of gadolinium europium ethylenediaminetetracetate salt.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS--

Please replace Page 8, line 1 as follows:

--I Claim:--

IN THE CLAIMS:

Please amend the following claims:

1. (Amended) An electroluminescent material which emits light in the ultra-violet region of the spectrum which comprises an organic metallic complex of at least one selected from the group consisting of a transition metal, a lanthanide and an actinide and a polyamine ligand.
2. (Amended) An electroluminescent material as claimed in claim 1 comprising gadolinium in the III state and a polyamine ligand.
3. (Amended) An electroluminescent material as claimed in claim 1 wherein the ligand is selected from the group consisting of ethylene diamine tetramine, DCTA, DTPA and TTHA.
4. (Amended) An electroluminescent material as claimed in claim 25 in which the complex is in the form of a salt.
6. (Amended) An electroluminescent material as claimed in claim 4 in which the salt is selected from the group consisting of transition metal, lanthanide and actinide salt.
7. (Amended) An electroluminescent material as claimed in claim 4 in which the salt has a formula $\text{Ln}^*[\text{Ln}(\text{EDTA})]_3$ and wherein Ln and Ln^* are the same or different and are selected from the group consisting of transition metals, lanthanides and actinides.

PATENT

8. (Amended) An electroluminescent material as claimed in claim 7 in which Ln and Ln* are selected from the group consisting of Gd, Sm, Eu, Tb and Dy.
9. (Amended) An electroluminescent material as claimed in claim 1 wherein the complex is Gd[Eu(EDTA)]₃.
10. (Amended) An electroluminescent device which comprises sequentially (i) a first electrode comprising a transparent conductive substrate (ii) a layer of a hole transmitting material (iii) a layer of an electroluminescent material which emits light in the ultra-violet region of the spectrum and which comprises an organic metallic complex of a transition metal, lanthanide or actinide and a polyamine ligand and (iv) a layer of an electron transmitting material and (v) a metal electrode.
11. (Amended) An electroluminescent device as claimed in claim 10 in which the transparent substrate is a conductive glass or plastic material which acts as the anode.

Please cancel claim 12.

13. (Amended) An electroluminescent device as claimed in claim 10 in which there is a hole transporting material mixed with the electroluminescent material in a ratio of 5 to 95% of the electroluminescent material to 95 to 5% of the hole transporting material.
14. (Amended) An electroluminescent device as claimed in claim 10 in which the hole transporting material is an aromatic amine complex.

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15. (Amended) An electroluminescent device as claimed in claim 10 in which the hole transporting material is selected from the group consisting of poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl)-1,1' -biphenyl -4,4'-diamine (TPD) and polyaniline.

Please cancel claims 16 and 17.

18. (Amended) An electroluminescent device as claimed in claim 10 in which an electron injecting material is mixed with the electroluminescent material and co-deposited with it.

19. (Amended) An electroluminescent device as claimed in claim 18 in which the electron injecting material is selected from the group consisting of a metal complex oxadiazole and an oxadiazole derivative.

20. (Amended) An electroluminescent device as claimed in claim 19 in which the electron injecting material is selected from the group consisting of an aluminum quinolate and 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4 oxadiazole.

22. (Amended) An electroluminescent device as claimed in claim 11 in which the anode is selected from the group consisting of aluminum alloy, magnesium alloy, lithium alloy, calcium alloy and magnesium silver alloy.

Please cancel claim 23.

24. (Amended) An electroluminescent device as claimed in claim 10 in which there is at least one layer which incorporates a dye which fluoresces in ultra-violet light to give emitted light in the colour spectrum.

Please add the following claims:

25. (New) An electroluminescent material as claimed in claim 1 wherein the organic metallic complex comprises gadolinium in the III state and a ligand selected from the group consisting of ethylene diamine tetramine, DCTA, DTPA and TTHA.

26. (New) An electroluminescent device as claimed in claim 10 in which the electroluminescent material is an organic metallic complex of gadolinium in the III state.

27. (New) An electroluminescent device as claimed in claim 10 in which the ligand is selected from the group consisting of ethylene diamine tetramine, DCTA, DTPA and TTHA.

28. (New) An electroluminescent device as claimed in claim 10 wherein the complex comprises gadolinium in the III state and ligand is selected from the group consisting of ethylene diamine tetramine, DCTA, DTPA and TTHA.

29. (New) An electroluminescent device as claimed in claim 28 in which the complex is in the form of a salt.

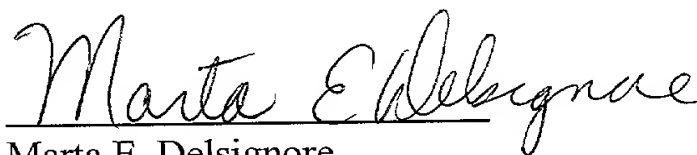
30. (New) An electroluminescent device as claimed in claim 28 in which the complex is in the form of an alkali metal salt.
31. (New) An electroluminescent device as claimed in claim 29 in which the salt is a salt of at least one selected from the group consisting of transition metal, lanthanide and actinide.
32. (New) An electroluminescent device as claimed in claim 10 in which the electroluminescent material is in the form of the salt of formula $\text{Ln}^*[\text{Ln}(\text{EDTA})]_3$ where Ln and Ln^* are the same or different and are selected from the group consisting of transition metals, lanthanides and actinides.
33. (New) An electroluminescent device as claimed in claim 32 in which Ln and Ln^* are selected from the group consisting of Gd, Sm, Eu, Tb and Dy.
34. (New) An electroluminescent device as claimed in claim 10 in which the metal complex is $\text{Gd}[\text{Eu}(\text{EDTA})]_3$.

REMARKS

By the foregoing amendment the specification and claims have been amended to conform more closely with U.S. patent practice.

Attached hereto is a marked-up version of the changes made to the specification and claims by the current amendment. The attached page is captioned "**Version with markings to show changes made.**"

Respectfully submitted,



Marta E. Delsignore
Patent Office Reg. No. 32,689

Attorney for Applicants

BAKER BOTTS L.L.P.
30 Rockefeller Plaza
New York, NY 10112
(212) 408-2632

VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE SPECIFICATION:

Page 1, before the first full paragraph please insert:

--This application is a national stage application of PCT/GB00/00268 which was published in English under publication number WO 00/44851 on August 3, 2001--

Page 1, after line 3, please insert:

--The present invention relates to electroluminescent materials which can emit light in the ultra-violet region of the spectrum and devices made using such materials.

BACKGROUND OF THE INVENTION--

Page 2, after line 13, please insert:

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SUMMARY OF THE INVENTION--

Page 4, after line 10, please insert:

--The materials of the present invention can be incorporated into electroluminescent devices which emit ultra-violet light and the invention include such electroluminescent devices.

BRIEF DESCRIPTION OF THE DRAWINGS

Figure 1 is a spectrum gadolinium ethylenediaminetetracetate sodium salt; and

Figure 2 is a spectrum of gadolinium europium ethylenediametetracetate salt.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS--

Please replace Page 8, line 1 as follows:

--[Claims] I Claim--

IN THE CLAIMS:

Please amend the following claims:

1. (Amended) An electroluminescent material which emits light in the ultra-violet region of the spectrum which comprises an organic metallic complex of at least one selected from the group consisting of a transition metal, a lanthanide [or] and an actinide and a polyamine ligand.
2. (Amended) An electroluminescent material as claimed in claim 1 [in which the metal is] comprising gadolinium in the III state and a polyamine ligand.
3. (Amended) An electroluminescent material as claimed in claim 1 [or 2 in which] wherein the ligand is selected from the group consisting of ethylene diamine tetramine, DCTA, DTPA [or] and TTHA.
4. (Amended) An electroluminescent material as claimed in claim [3] 25 in which the complex is in the form of a salt.

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6. (Amended) An electroluminescent material as claimed in claim 4 in which the salt is [a] selected from the group consisting of transition metal, lanthanide [or] and actinide salt.
7. (Amended) An electroluminescent material as claimed in claim 4 in which the salt [is] has a formula $\text{Ln}^*[\text{Ln}(\text{EDTA})]_3$ and wherein Ln and Ln^* are the same or different and are selected from the group consisting of transition metals, lanthanides and actinides.
8. (Amended) An electroluminescent material as claimed in claim 7 in which Ln and Ln^* [is] are selected from the group consisting of Gd, Sm, Eu, Tb and Dy.
9. (Amended) An electroluminescent material as claimed in [any one of claims 1, 4 or 5 in which] claim 1 wherein the [metal] complex is $\text{Gd}[\text{Eu}(\text{EDTA})]_3$.
10. (Amended) An electroluminescent device which comprises [a transparent substrate on which is deposited an electroluminescent material as claimed in any one of the preceding claims] sequentially (i) a first electrode comprising a transparent conductive substrate (ii) a layer of a hole transmitting material (iii) a layer of an electroluminescent material which emits light in the ultra-violet region of the spectrum and which comprises an organic metallic complex of a transition metal, lanthanide or actinide and a polyamine ligand and (iv) a layer of an electron transmitting material and (v) a metal electrode.
11. (Amended) An electroluminescent device as claimed in [claims 1 to] claim 10 in which the transparent substrate is a conductive glass or plastic material which acts as the anode.

Please cancel claim 12.

13. (Amended) An electroluminescent device as claimed in [any one of claims] claim 10 [to 12] in which there is a hole transporting material mixed with the electroluminescent material in a ratio of 5 to 95% of the electroluminescent material to 95 to 5% of the hole transporting [compound] material.

14. (Amended) An electroluminescent device as claimed in claim [12 or 13] 10 in which the hole transporting [layer] material is an aromatic amine complex.

15. (Amended) An electroluminescent device as claimed in claim [14] 10 in which the hole transporting [layer] material is selected from the group consisting of poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl)-1,1' -biphenyl -4,4'-diamine (TPD) [or] and polyaniline.

Please cancel claims 16 and 17.

18. (Amended) An electroluminescent device as claimed in [any one of claims] claim 10 [to 17] in which an electron injecting material is mixed with the electroluminescent material and co-deposited with it.

19. (Amended) An electroluminescent device as claimed in claim [17 or] 18 in which the electron injecting material is selected from the group consisting of a metal complex [or] oxadiazole [or] and an oxadiazole derivative.

PATENT

20. (Amended) An electroluminescent device as claimed in claim 19 in which the electron injecting material is selected from the group consisting of an aluminum quinolate [or] and 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4 oxadiazole.

22. (Amended) An electroluminescent device as claimed in claim [21] 11 in which the anode is selected from the group consisting of [a] aluminum alloy, magnesium alloy, lithium alloy, calcium alloy [or a] and magnesium silver alloy.

Please cancel claim 23.

24. (Amended) An electroluminescent device as claimed in [any one of the preceding claims] claim 10 [to 23] in which there is [a] at least one layer [or layers] which incorporates a dye which fluoresces in ultra-violet light to give emitted light in the colour spectrum.

2 / PRTS

JC17 Rec'd PCT/PTO 30 JUL 2001

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WO 00/44851

PCT/GB00/00268

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Electroluminescent Materials

The present invention relates to electroluminescent materials which can emit light in the ultra-violet region of the spectrum and devices made using such materials.

5 Materials which emit light when an electric current is passed through them are well known and used in a wide range of display applications and organic polymers have been proposed as useful in electroluminescent devices, but using these polymers it is not possible to obtain pure colours, they are expensive to make and have a relatively low efficiency.

10

Another compound which has been proposed is aluminium quinolate, but this requires dopants to be used to obtain a range of colours and has a relatively low efficiency.

15

In an article in Chemistry letters pp 657-660, 1990 Kido et al disclosed that a terbium (III) acetyl acetonate complex was green electroluminescent and in an article in Applied Physics letters 65 (17) 24 October 1994 Kido et al disclosed that a europium (III) triphenylene diamine complexes was red electroluminescent but these were unstable in atmospheric conditions and difficult to produce as films.

20

The complexes disclosed in these articles had a relatively low photoluminescent efficiency and were only able to produce green or red light and other colours could not be produced.

25

Patent Applications WO 98/58037 and WO 98/55561 disclose electroluminescent materials which emit light in the visible spectrum and disclose electroluminescent devices incorporating these materials.

30

The wavelength of the emitted light from an organo metallic complex which is an electroluminescent material depends on the metal or metals and the ligands.

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In general the shorter the wavelength of the emitted light the harder it is to obtain effective electroluminescent materials and in particular electroluminescent materials which emit light in the ultra-violet region of the spectrum have hitherto not been possible to produce.

5

An electroluminescent material which can emit ultra-violet light would have a range of applications and would enable there to be a source of ultra-violet light without the need for high temperature or complex high energy or vacuum equipment and so could replace such sources of ultra-violet light.

10

For example, there are devices and displays etc. where ultra-violet light is used to excite other materials to cause these other materials to fluoresce in the visible spectrum.

15

We have now devised organo-metallic complexes which can emit ultra-violet light.

According to the invention there is provided an electroluminescent material which emits light in the ultra-violet region of the spectrum which comprises an organic metallic complex of a transition metal, lanthanide or actinide and a polyamine ligand.

20

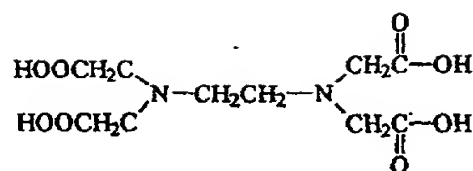
The preferred metal is gadolinium in the III state.

Preferred ligands are ethylene diamine tetramine EDTA, DCTA, DTPA and TTHA.

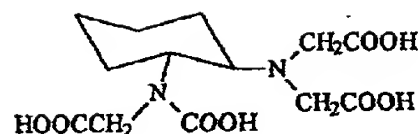
25

The structural formulae of these compounds in the acetic acid form are shown below.

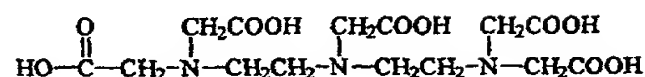
- 3 -



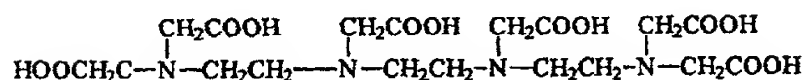
EDTA



DCTA

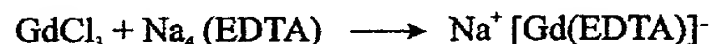


DTPA



TTHA

- 5 The organo metallic complexes can be made by the reaction of gadolinium chloride with an alkali metal salt of the amine e.g. the sodium salt, for EDTA the reaction is



For the other polyamine the corresponding alkali metal salt is used.

- 10 With polyamines which are more than tri-functional e.g. EDTA, DCTA, DTPA, TTHA the complexes can be in the form of a salt e.g. an alkali metal salt and can be used in this form. Alternatively a transition metal, lanthanide or actinide salt e.g. $\text{Ln}^*[\text{Ln}(\text{EDTA})]_3$ where Ln and Ln^* is a transition metal, lanthanide or actinide and preferably a lanthanide e.g. Gd, Sm, Eu, Tb, Dy, etc.

15

Particularly preferred mixed complexes are the gadolinium complexes e.g. $\text{Gd}[\text{Eu}(\text{EDTA})]_3$.

- 4 -

It has surprisingly been found that the lanthanide salts of the organo-metallic complexes exhibit a different electroluminescent spectrum than mixed organo-metallic complexes.

- 5 The mixed lanthanide salts can be made by reacting a lanthanide chloride with a lanthanide polyamine complex.

10 The materials of the present invention can be incorporated into electroluminescent devices which emit ultra-violet light and the invention include such electroluminescent devices.

15 The ultra-violet light emitting devices of the invention comprise a transparent substrate which is a conductive glass or plastic material which acts as the anode, preferred substrates are conductive glasses such as indium tin oxide coated glass, but any glass which is conductive or has a conductive layer can be used. Conductive polymers and conductive polymer coated glass or plastics materials can also be used as the substrate. The electroluminescent material can be deposited on the substrate directly by evaporation from a solution of the material in an organic solvent. The solvent which is used will depend the material for example alcohols such as ethanol, ketones such as acetone and methyl acetylacetonate and chlorinated hydrocarbons such as dichloromethane are suitable in many cases.

20

Alternatively the material can be deposited by spin coating or by vacuum deposition from the solid state e.g. by sputtering or any other conventional method can be used.

25

In one embodiment there is a hole transporting layer deposited on the transparent substrate and the electroluminescent material is deposited on the hole transporting layer. The hole transporting layer serves to transport; holes and to block the electrons, thus preventing electrons from moving into the electrode without

- 5 -

recombining with holes. The recombination of carriers therefore mainly takes place in the emitter layer.

5 Hole transporting layers are used in polymer electroluminescent devices and any of the known hole transporting materials in film form can be used.

10 The hole transporting layer can be made of a film of an aromatic amine complex such as poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl)-1,1'-biphenyl -4,4'-diamine (TPD), polyaniline etc.

15 Optionally dyes such as fluorescent laser dyes, luminescent laser dyes can be included so that these dyes fluoresce in the ultra-violet light to give emitted light of a particular colour spectrum.

20 Preferably the electroluminescent material is mixed with a polymeric material such as a polyolefin e.g. polyethylene, polypropylene etc. and preferably polystyrene. Preferred amounts of active material in the mixture is from 95% to 5% by weight of active material and more preferably 25 to 20% by weight.

25 The hole transporting material can optionally be mixed with the electroluminescent material in a ratio of 5-95% of the electroluminescent material to 95 to 5% of the hole transporting compound. In another embodiment of the invention there is a layer of an electron injecting material between the cathode and the electroluminescent material layer, this electron injecting material is preferably a metal complex such as a metal quinolate e.g. an aluminium quinolate which will transport electrons when an electric current is passed through it. Alternatively the electron injecting material can be mixed with the electroluminescent material and co-deposited with it.

30 In a preferred structure there is a substrate formed of a transparent conductive material which is the anode on which is successively deposited a hole transportation

- 6 -

layer, the electroluminescent material layer and an electron injection layer which is connected to the anode. The anode can be any low work function metal e.g. aluminium, calcium, lithium, silver/magnesium alloys etc.,

- 5 There can be a layer or layers containing a fluorescent material in the device so that the ultra-violet light emitted will cause the material to fluoresce and emit light of a particular colour spectrum.

The invention is described in the following examples.

10

Example 1

Gadolinium ethylenediaminetetraacetate sodium salt, Na[Gd(EDTA)]

Gadolinium chloride (10 mmol) was dissolved in water (5 ml). Ethylenediaminetetraacetic acid, tetrasodium salt hydrate (10 mmol) was dissolved in water (10 ml) and added portionwise to the gadolinium chloride solution. The solution became warm, and after ca. 15 minutes, a white precipitate was formed. The mixture was left for a further 2 hours. The precipitate was filtered off to give a white solid which was washed with water (2 x 5 ml) and dried in air to yield Gadolinium ethylenediaminetetraacetate sodium salt, Na[Gd(EDTA)]

20

Example 2

Gadolinium europium ethylenediaminetetraacetate salts, Gd[Eu(EDTA)]₃

Europium ethylenediaminetetraacetate sodium salt Na[Eu(EDTA)] (4.5 mmol) was dissolved in methanol (30 ml). Gadolinium chloride (1.5 mmol) was dissolved in water (5 ml) and added portionwise to the europium ethylenediaminetetraacetate sodium salt solution. A white precipitate was immediately formed. The mixture was left for a further 1 hour. The precipitate was filtered off to give a white solid which was washed with methanol (2 x 10 ml) and dried in air to yield the product Gadolinium europium ethylenediaminetetraacetate salts, Gd[Eu(EDTA)]₃.

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- Photoluminescence was excited using 325nm line of Liconix 4207 NB, He/Cd laser. The laser power incident at the sample (0.3mWcm^{-2}) was measured by a Liconix 55PM laser power meter. The radiance calibration was carried out using Bentham radiance standard (Bentham SRS8, Lamp current 4,000A, calibrated by National Physical laboratories, England. The PL studies were carried out on samples or films. The Complexes of the examples were tested and the results shown in the Spectra attached as Figs. 1 and 2.

- An electroluminescent device constructed with a film of the compounds of examples 1 and 2 with an indium coated glass as anode and an aluminium cathode emitted light in the ultra-violet spectrum when a low voltage was applied across the film. The light was emitted in the same spectrum ranges as in figs. 1 and 2

Claims

1. An electroluminescent material which emits light in the ultra-violet region of the spectrum which comprises an organic metallic complex of a transition metal,
5 lanthanide or actinide and a polyamine ligand.
2. An electroluminescent material as claimed in claim 1 in which the metal is gadolinium in the III state.
- 10 3. An electroluminescent material as claimed in claim 1 or 2 in which the ligand is ethylene diamine tetramine, DCTA, DTPA or TTHA.
4. An electroluminescent material as claimed in claim 3 in which the complex is in the form of a salt .
15
5. An electroluminescent material as claimed in claim 4 in which the complex is in the form of an alkali metal salt
6. An electroluminescent material as claimed in claim 4 in which the salt is a
20 transition metal, lanthanide or actinide salt
7. An electroluminescent material as claimed in claim 4 in which the salt is $\text{Ln}^*[\text{Ln}(\text{EDTA})]_3$ where Ln and Ln^* is selected from transition metals, lanthanides and actinides.
25
8. An electroluminescent material as claimed in claim 7 in which Ln and Ln^* is selected from Gd, Sm, Eu, Tb, Dy.
9. An electroluminescent material as claimed in any one of claims 1, 4 or 5 in which
30 the metal complex is $\text{Gd}[\text{Eu}(\text{EDTA})]_3$.

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10. An electroluminescent device which comprises a transparent substrate on which is deposited an electroluminescent material as claimed in any one of the preceding claims.

5 11. An electroluminescent device as claimed in claims 1 to 10 in which the transparent substrate is a conductive glass or plastic material which acts as the anode.

10 12. An electroluminescent device as claimed in any one of claims 10 to 11 in which there is a hole transporting layer deposited on the transparent substrate and the electroluminescent material is deposited on the hole transporting layer.

15 13. An electroluminescent device as claimed in any one of claims 10 to 12 in which there is a hole transporting material mixed with the electroluminescent material in a ratio of 5 to 95% of the electroluminescent material to 95 to 5% of the hole transporting compound.

14. An electroluminescent device as claimed in claim 12 or 13 in which the hole transporting layer is an aromatic amine complex.

20 15. An electroluminescent device as claimed in claim 14 in which the hole transporting layer is poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl) -1,1' -biphenyl -4,4'-diamine (TPD) or polyaniline.

25 16. An electroluminescent device as claimed in any one of claims 11 to 15 in which there is a metal anode in contact with the electroluminescent material.

30 17. An electroluminescent device as claimed in any one of claims 10 to 16 in which there is a layer of an electron injecting material between the cathode and the electroluminescent material layer

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18. An electroluminescent device as claimed in any one of claims 10 to 17 in which an electron injecting material is mixed with the electroluminescent material and co-deposited with it.

5 19. An electroluminescent device as claimed in claim 17 or 18 in which the electron injecting material is a metal complex or oxadiazole or an oxadiazole derivative.

10 20. An electroluminescent device as claimed in claim 19 in which the electron injecting material is an aluminium quinolate or 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4 oxadiazole.

21. An electroluminescent device as claimed in any one of the preceding claims 10 to 20 in which the anode is a metal.

15 22. An electroluminescent device as claimed in claim 21 in which the anode is a aluminium, magnesium, lithium, calcium or a magnesium silver alloy.

20 23. An electroluminescent device as claimed in any one of the preceding claims 10 to 22 in which there are a plurality of layers of electroluminescent material.

25 24. An electroluminescent device as claimed in any one of the preceding claims 10 to 23 in which there is a layer or layers which incorporates a dye which fluoresces in ultra-violet light to give emitted light in the colour spectrum.

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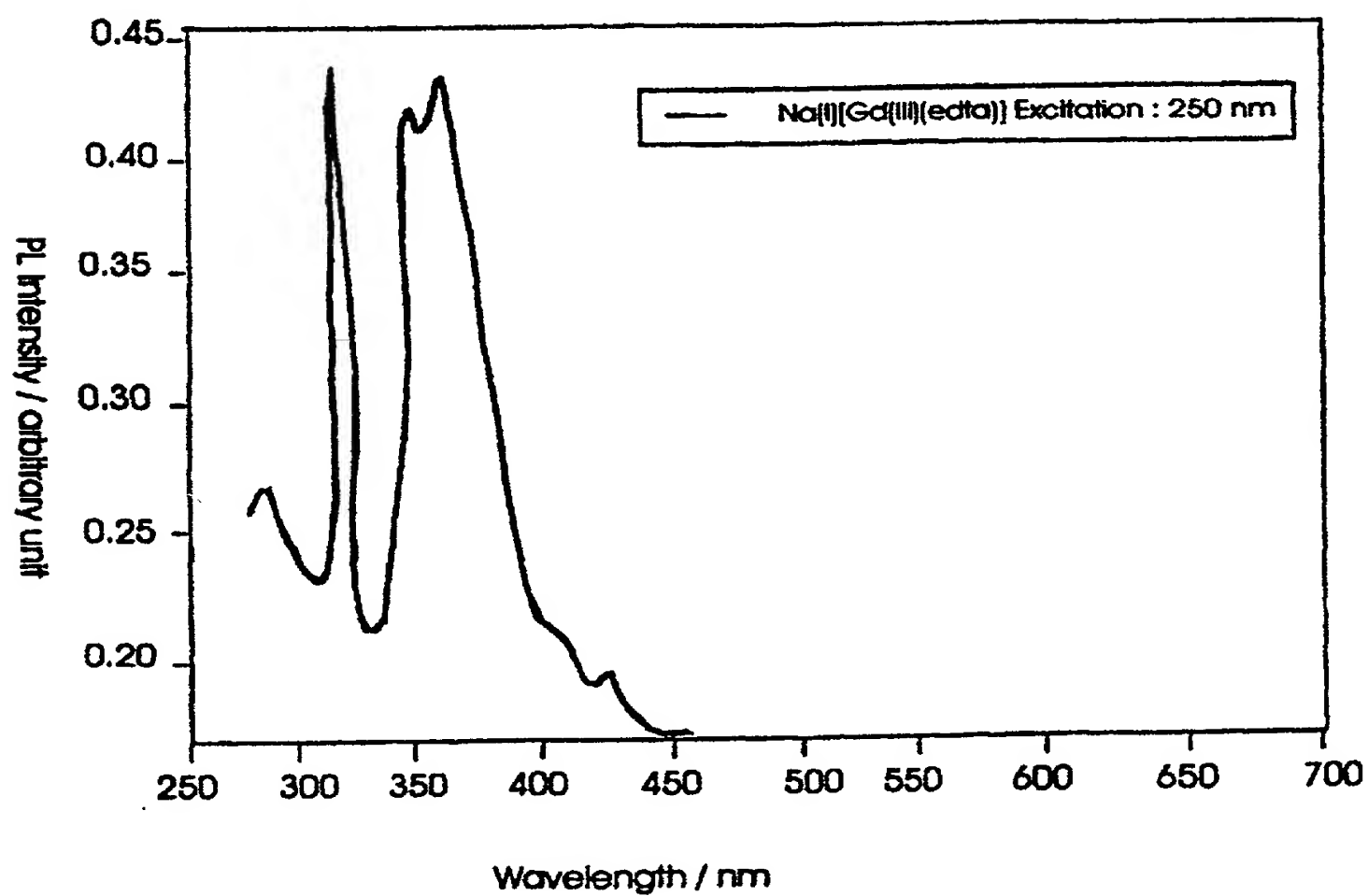


Fig. 1

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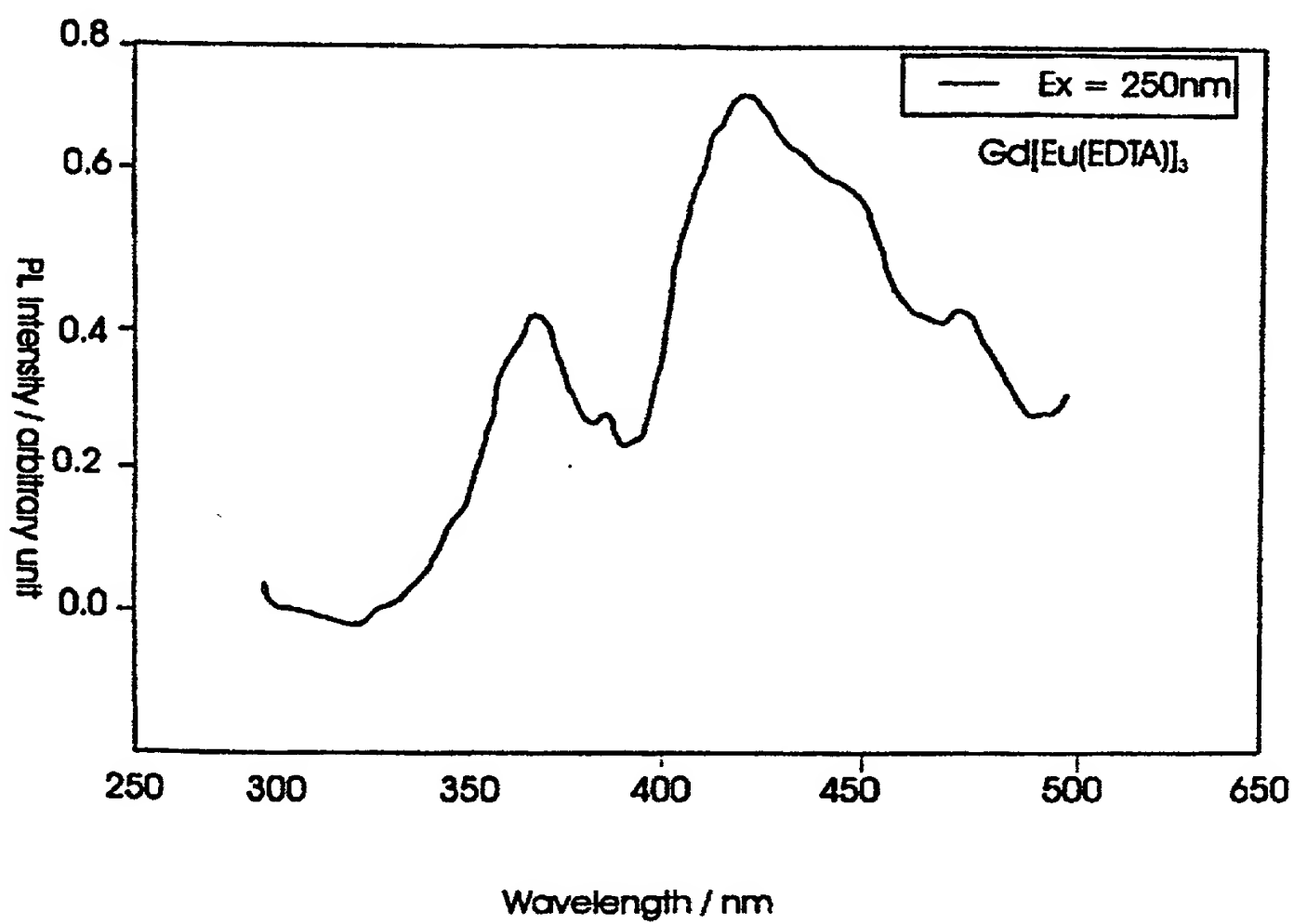


Fig. 2

Docket No.
UEL,017-US

Declaration and Power of Attorney For Patent Application

English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

ELECTROLUMINESCENT MATERIALS

the specification of which

(check one)

☐ is attached hereto.

☒ was filed on 31st January 2000 as United States Application No. or PCT International Application Number PCT/GB00/00268 and was amended on _____

(if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119(a)-(d) or Section 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)

Priority Not Claimed

<u>UK 99 01971.3</u>	<u>UK</u>	<u>1st February 1999</u>	<input type="checkbox"/>
(Number)	(Country)	(Day/Month/Year Filed)	
<u> </u>	<u> </u>	<u> </u>	<input type="checkbox"/>
(Number)	(Country)	(Day/Month/Year Filed)	
<u> </u>	<u> </u>	<u> </u>	<input type="checkbox"/>
(Number)	(Country)	(Day/Month/Year Filed)	

I hereby claim the benefit under 35 U.S.C. Section 119(e) of any United States provisional application(s) listed below:

(Application Serial No.)

(Filing Date)

(Application Serial No.)

(Filing Date)

(Application Serial No.)

(Filing Date)

I hereby claim the benefit under 35 U. S. C. Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. Section 112, I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, C. F. R., Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application:

(Application Serial No.)

(Filing Date)

(Status)
(patented, pending, abandoned)

(Application Serial No.)

(Filing Date)

(Status)
(patented, pending, abandoned)

(Application Serial No.)

(Filing Date)

(Status)
(patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. *(list name and registration number)*

Marta E. Delsignore #32689

Send Correspondence to: Marta E. Delsignore
Baker Botts L.L.C.
30 Rockefeller Plaza
New York, NY 10112-0228

Direct Telephone Calls to: *(name and telephone number)*
Marta E. Delsignore (212) 408 2632

Full name of sole or first inventor Poopathy KATHIRGAMANATHAN	
Sole or first inventor's signature <i>[Signature]</i>	Date 23-04-01
Residence North Harrow, Middlesex, United Kingdom GBX	
Citizenship UK	
Post Office Address 14 Sandhurst Avenue,	
North Harrow, Middlesex, United Kingdom	

Full name of second inventor, if any	
Second inventor's signature	Date
Residence	
Citizenship	
Post Office Address	